09/914866 JC03 Rec'd PCT/FTO 04 SEP 2001

JORDAN AND HAMBURG LLP

122 East 42nd Street, New York, NY 10168

Certificate of Express Mailing Under 37 CFR 1.10

Tel: (212) 986-2340 Fax: (212) 953-7733

Customer No.: 000028107

Docket No.: F-7104

Filing Date: September 4, 2001

"Express Mail" label number: EL 666236343US Date of Deposit: 9/4/01
I hereby certify that this correspondence is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 in an envelope addressed to ASSISTANT COMMISSIONER OF PATENTS, WASHINGTON, DC 20231 having the above "Express Mail" label number on the date of deposit indicated above.
Madeline Gonzalez Madeline Herale
(Typed or printed name of person mailing paper or fee) (Signature of person mailing paper or fee)
THE ASSISTANT COMMISSIONER FOR PATENTS Washington, D. C. 20231
[-] ATTN: BOX PATENT APPLICATION
[] ATTN: BOX DESIGN PATENT APPLICATION
[x] ATTN: BOX PCT
[x] THIS IS THE 35 U.S.C 371 NATIONAL STAGE OF PCT/EP00/01438 FILED
<u>February 22, 2000</u>
Sir: Transmitted herewith for filing is the [X] Utility [] Design nonprovisional patent application of:
Inventor / Application Identifier: Michael ROREGER, et al.
[X] See Inventor Information Sheet attached
For: METHOD FOR PRODUCING A FLAT PREPARATION WITH AT LEAST ONE SINGLE-LAYER INGREDIENT MATRIX
[] This is a new patent application. [X] This is the 35 U.S.C. 371 National Stage Application of the above-identified PCT Application. [] This is a: [] Continuation Application [] Divisional Application [] Continuation-in-Part Application of prior Application Serial No
[] Cancel in this application original claims of the prior application before calculating the filing fee.
[] Amend the specification by inserting before the first line the sentence: This is a [] Continuation, [] Division, [] Continuation-in-part, of Application
[] Incorporation By Reference. The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied, is considered as being part of the disclosure of the accompanying

application and is hereby incorporated by reference therein.

ENCL	OSED .	ARE THE FOLLOWING:
Х	2	Sheets of drawings ([x] formal [] informal size A4).
Х	18	Pages of specification including abstract and claims.
Х	20	Total pages.
	Con	nbined Declaration and Power of Attorney
		Newly executed
		Copy from prior application
		Inventors deleted; see attached statement
	Sequ	ience Listing
		Computer Readable Copy
		Paper copy
		Statement verifying identity of above copies
Х	Retu	rn Receipt Postcard
х	Preli	minary Amendment
	Assig	gnment to:
		Assignment is of record in prior application Serial No
		Assignment Recordation Form Cover Sheet.
		Charge \$40.00 to Deposit Account No. 10-1250 for recording Assignment.
х	Inforr	nation Disclosure Statement
х	Inform	nation Disclosure Citation
	Englis	sh translation
Х	Appli	cation Data Sheet

have been filed in the International Bureau

were filed in prior application:

09/914866 JC03 Record Full 10 0 4 SEP 2001

X

CLAIMS FILED AND FILING FEI ITEM	E CALCUL	ATION		Rate	Applied
[] Base Fee - Non PCT				\$710	Fee
Base Fee - PCT IPEA-US			\$690		
Base Fee - PCT ISA-US			\$710		
[] Base Fee - PCT not ISA or IPEA				\$1,000	
[X] Base Fee - PCT with EPO or JPO Search Report				\$860	\$860
[] Base Fee - Design				\$320	
Claim Fees	Number Filed	Base Number	Number Extra over Base		
Total Claims	34	20	14	\$18	\$252
Independent Claims	1	3	0	\$80	\$0
Multiple Dependent Claim Fee	-	•		\$270	\$270
[] Small Entity Status is Asserted					(\$0)
[] Foreign Language Filing Fee				\$130	\$0
TOTAL FILING FEE				_l	\$1,382

0 4 SEP 2001

- [X] Please charge Deposit Account No. 10-1250 in the amount of the above TOTAL FILING FEE. A duplicate copy of this sheet is attached.
- [X] Please charge to Deposit Account No. 10-1250 any further fees under: 37 CFR 1.16; 37 CFR 1.17; and 37 CFR 1.492.

JORDAN AND HAMBURG LLP

C. Bruce Hamburg

Reg. No. 22389 Attorney for Applicant

INVENTOR INFORMATION SHEET

Docket Number: F-7104

Title: METHOD FOR PRODUCING A FLAT PREPARATION WITH AT LEAST ONE

SINGLE-LAYER INGREDIENT MATRIX

Filing Date: 9/4/01

1. Full Name of Inventor Michael ROREGER	Family Name ROREGER	First Given Name Michael	Second Given Name
Citzenship Germany	Residence City Neuwied	Residence State or Province	Residence Country Germany
Postal Address Pestalozzi-Strasse 38	City Neuwied	Province/State	Postal Code/Country D-56567 Germany
2. Full Name of Inventor Malgorzata KLOCZKO	Family Name KLOCZKO	First Given Name Malgorzata	Second Given Name
Citzenship Poland	Residence City Linz	Residence State or Province	Residence Country Germany
Postal Address Magdalena- Daemen-Strasse 18	City Linz	Province/State	Postal Code/Country D-53545 Germany

Application Data Sheet

Application Information

Application Type::

Regular

Subject Matter::

Utility

Suggested Group Art Unit::

Sequence submission?::

Computer Readable Form

(CRF)?::

Title::

METHOD FOR PRODUCING A FLAT

PREPARATION WITH AT LEAST ONE

SINGLE-LAYER INGREDIENT MATRIX

Attorney Docket Number::

F-7104

Suggested Drawing Figure::

Total Drawing Sheets::

1

Small Entity::

No

Applicant Information

Applicant Authority Type::

Inventor

Primary Citizenship Country:: Germany

Status::

Full Capacity

Given Name::

Michael

Middle Name::

Family Name::

ROREGER

Page # 1 Initial 9/4/01

The second second (1) 2 m City of Residence::

Neuwied

State or Province of

Residence::

Country of Residence:: Germany

Street of Mailing Address:: Pestalozzi-Strasse 38

City of Mailing Address:: Neuwied

State or Province of Mailing

Address::

Country of Mailing Address:: Germany

Postal or Zip Code of

Mailing Address::

D-56567

Applicant Authority Type:: Inventor

Primary Citizenship Country:: Poland

Status::

Full Capacity

Given Name::

Malgorzata

Middle Name::

Family Name::

KLOCZKO

City of Residence::

Linz

State or Province of

Residence::

Country of Residence::

Germany

Page # 2 Initial 9/4/01

Street of Mailing Address:: Magdalena-Daemen-Strasse 18

City of Mailing Address::

Linz

State or Province of Mailing

Address::

Country of Mailing Address:: Germany

Postal or Zip Code of

Mailing Address::

D-53545

Correspondence Information

Correspondence Customer

Number::

000028107

Representative Information

Representative Designation::	Registration number::	Name::
Primary	22389	C. Bruce Hamburg

Domestic Priority Information

Application::	Continuity Type::	Parent Application::	Parent Filing Date::
This application	National Stage of	PCT/EP00/01438	02/22/00

Foreign Priority Information

Country::	Application Number::	Filing Date::	Priority Claimed::
Germany	19909493.4	03/04/99	Yes

09/914866 04 SEP 2001

F-7104

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant

Michael BOREGER et al.

Serial No.

Filing concurrently with application

Filed

Concurrently

For

METHOD FOR PRODUCING A FLAT

PREPARATION WITH AT LEAST ONE SINGLE-

LAYER INGREDIENT MATRIX

Group Art Unit

(Not yet known)

Examiner

(Not yet known)

Assistant Commissioner for Patents Washington, D.C. 20231

PRELIMINARY AMENDMENT

Sir:

Preliminary to examination, please amend the above-identified patent application as follows:

IN THE TITLE:

Change the title to --METHOD FOR PRODUCING A FLAT PREPARATION WITH AT LEAST ONE SINGLE-LAYER INGREDIENT MATRIX--.

IN THE CLAIMS:

Please amend claims 1-15 as follows. Appendix I is attached hereto having marked versions of said claims with amendments indicated by brackets and underlining.

- 1. (amended) Process for producing a sheetlike article comprising a single-layer homogenous matrix containing at least one active substance for controlled release of the at least one active substance to the vicinity of an application site, the at least one active substance being selected from crop protection agents, biocides, fertilizers, plant strengtheners, cosmetic active principles and fragrances, comprising the following temporally and spatially separate steps:
- a) application of the at least one active substance to at least one of two layers, identical in composition, of a base material,
- b) placement of the two base material layers atop one another so as to enclose the at least one active substance applied, and irreversible joining of the layers with the at least one active substance therebetween under pressure to form a laminate, and
- c) storage of the laminate for predeterminable duration under defined conditions to effect migration of the at least one active substance into the base material layers and connection of the base material layers at their interfaces to form a single-layer homogenous matrix in which the at least one active substance is substantially uniformly distributed.

F-7104

- 2. (amended) Process according to Claim 1, wherein the at least one active substance is applied in step a) at a pressure ≤ 12 bar with metering.
- 3. (amended) Process according to Claim 1 or 2, wherein the pressure in step b) is from 2 to 10 bar.
- 4. (amended) Process according to Claim 1 or 2, wherein the storage of step c) is at a temperature of 15 to 30°C and the duration of the storage is at least 48 hours.
- 5. (amended) Process according to Claim 1 or 2, wherein the at least one active substance applied in step a) is in the form of a flowable medium having a viscosity of at least 1000 mPa.s.
- 6. (amended) Process according to Claim 5, wherein the at least one active substance applied in step a) contains auxiliaries.
- 7. (amended) Process according to Claim 1 or 2, wherein the application of the at least one active substance in step a) is continuous or intermittent.
- 8. (amended) Process according to Claim 1 or 2, wherein said matrix is self-adhesive.

- 9. (amended) Process according to Claim 1 or 2, wherein the at least one active substance is volatile or thermally labile.
- 10. (amended) Process according to Claim 1 or 2, wherein the base material comprises at least one polymer selected from the group consisting of ethylene-vinyl acetate copolymer, styrene/butadiene/styrene block copolymer, styrene/isoprene/styrene block copolymer, polyisobutylene, polyacrylates, polymethacrylates, polyvinyl esters, polyamide, polyesters, cellulose derivatives and silicones.
- 11. (amended) Process according to Claim 1 or 2, wherein the at least one active substance is in admixture with a tackifying substance.
- 12. (amended) Process according to Claim 1 or 2, wherein the at least one active substance comprises a mixture of Z,E-9,12-tetradecadienol and Z,E-9,12-tetradecadien-1-yl acetate.
- 13. (amended) Process according to Claim 1 or 2, wherein the base material contains at least one of said active substances.

F-7104

14. (amended) Process according to Claim 1 or 2, wherein the at least one active substance is at least one of dimethoate, imidacloprid, fenpropidine, acephate and acetamiprid.

15. (amended) Process according to Claim 1 or 2, wherein in step a) the at least one active substance is applied to at least one of the layers as a pattern.

Add the following claims:

- -- 16. Process according to Claim 3, wherein the pressure in step b) is from 3 to 5 bar.
- 17. Process according to Claim 4, wherein said temperature is 20 to 24°C.
- 18. Process according to Claim 15, wherein the pattern comprises stripes. -

IN THE ABSTRACT:

Please replace the abstract with the substitute abstract submitted on the following separate page.

ABSTRACT

A sheetlike article comprising a single-layer homogenous matrix containing at least one active substance such as a crop protection agent, biocide, fertilizer, plant strengthener, cosmetic substance or fragrance, is produced by applying the at least one active substance to at least one of two layers of identical composition, placing the two layers atop one another so as to enclose the at least one active substance, irreversibly joining the layers with the at least one active substance therebetween under pressure to form a laminate and storing the laminate under defined conditions for a duration sufficient to effect migration of the at least one active substance into the base layer and connection of the layers at their interfaces.

REMARKS

The foregoing makes formal changes in the claims to better adapt them to U.S. practice, provides an abstract in such paragraph format, not exceeding 150 words as required by present U.S. practice and conforms the title to the published PCT application.

Respectfully submitted,

JORDAN AND HAMBURG LLP

C. Bruce Hamburg Reg. No. 22,389

Attorney for Applicants

122 East 42nd Street New York, New York 10168 (212) 986-2340

APPENDIX I

AMENDED CLAIMS WITH AMENDMENTS INDICATED THEREIN BY BRACKETS AND UNDERLINING

- 1. (amended) Process for producing a sheetlike [formulation] <u>article</u> comprising [at least one] <u>a</u> single-layer [active substance] <u>homogenous</u> matrix <u>containing at least one active substance</u> for [the] controlled release of <u>the at least one</u> active substance to the vicinity of <u>an</u> application site, the <u>at least one</u> active [substances] <u>substance</u> being selected from crop protection agents, biocides, fertilizers, plant strengtheners, cosmetic active principles and fragrances, [characterized in that the matrix is produced using at least two layers (1, 2), identical in composition, of a base material in] <u>comprising</u> the following temporally and spatially separate steps:
- [a) provision of two prefabricable layers (1,2), identical in composition, of a base material,]
- [b)] <u>a)</u> application of <u>the at least one</u> active substance to at least one of [the] two layers [(1,2)], identical in composition, of a base material,
- [c)] b) placement of the two <u>base material</u> layers [(1, 2)] atop one another so as to enclose the <u>at least one</u> active substance applied, and irreversible joining <u>of the</u> <u>layers with the at least one active substance therebetween</u> under pressure to form a laminate, <u>and</u>
- [d)] c) storage of the laminate for predeterminable duration under defined conditions [, with] to effect migration of the at least one active substance into the base material layers [(1, 2)] and connection of the base material layers at their interfaces to form a single-layer homogenous matrix [featuring] in which the at least one active substance is substantially [uniform dispensation of active substance] uniformly distributed.

- 2. (amended) Process according to Claim 1, [characterized in that volumetric metering of] wherein the at least one active substance [medium] is [performed by admitting it] applied in step a) at a pressure ≤ 12 bar with metering.
- 3. (amended) Process according to Claim 1 or 2, [characterized in that the joining of the base material layers (1, 2) is performed under a laminating] wherein the pressure [of between] in step b) is from 2 [and] to 10 bar [, preferably between 3 and 5 bar].
- 4. (amended) Process according to [one or more of Claims] <u>Claim 1</u> [to 3] <u>or 2</u>, [<u>characterized</u> in that] <u>wherein</u> the storage of [the active substance matrix (14) is performed] <u>step c</u>) is at a temperature of [between] 15 [and] <u>to 30°C</u> [, preferably between 20-24°C, with a] and the duration of the storage is at least 48 hours.
- 5. (amended) Process according to [one or more of Claims] Claim 1 [to 4] or 2, [characterized in that] wherein the at least one active substance [is] applied in step a) is in the form of a flowable medium having a viscosity of at least 1000 mPa.s.
- 6. (amended) Process according to Claim 5, [characterized in that] wherein the at least one active substance [medium is] applied in step a) contains [with a fraction of] auxiliaries.
- 7. (amended) Process according to [one or more of Claims] <u>Claim 1 [to 6] or 2</u>, [<u>characterized</u> in that] <u>wherein</u> the application of <u>the at least one</u> active substance <u>in step a</u>) [performed continuously or intermittently] is <u>continuous or intermittent</u>.

- 8. (amended) Process according to [one or more of Claims] <u>Claim</u> 1 [to 7] <u>or 2</u>, [<u>characterized</u> in that the active substance] <u>wherein said</u> matrix [(14)] is [made] self-adhesive.
- 9. (amended) Process according to [one or more of Claims] <u>Claim 1</u> [to 8] <u>or 2</u>, [<u>characterized</u> in that] <u>wherein the at least one active substance is volatile or thermally labile [active substances are incorporated between the base material layers (1, 2) of the matrix].</u>
- 10. (amended) Process according to [one or more of Claims] Claim 1 [to 9] or 2, [characterized in that] wherein the base material comprises at least one polymer [from one of the following groups is] selected [as the matrix base material:] from the group consisting of ethylene-vinyl acetate copolymer, [block copolymer, e.g.] styrene/butadiene/styrene [or] block copolymer, styrene/isoprene/styrene block copolymer, polyisobutylene, polyacrylates, polymethacrylates, polyvinyl esters, polyamide, polyesters, cellulose derivatives and silicones.
- 11. (amended) Process according to [one or more of Claims] <u>Claim 1 [to 10] or 2</u>, [<u>characterized</u> in that an] <u>wherein the at least one active substance [medium] is [used which comprises] in admixture with a tackifying [substances] <u>substance</u>.</u>
- 12. (amended) Process according to [one or more of Claims] <u>Claim 1 [to 11] or 2</u>, [<u>characterized</u> in that an] <u>wherein the at least one</u> active substance [is used which is] <u>comprises</u> a mixture of Z,E-9,12-tetradecadienol and Z,E-9,12-tetradecadien-1-yl acetate.

F-7104

- 13. (amended) Process according to [one or more of Claims] <u>Claim 1 [to 12] or 2</u>, [<u>characterized</u> in that] <u>wherein</u> the [matrix layers comprise one or more] <u>base</u> <u>material contains at least one of said</u> active substances.
- 14. (amended) Process according to [one or more of Claims] <u>Claim 1 [to 13] or 2</u>, [<u>characterized</u> in that] <u>wherein the</u> at least one [of the] active [substances] <u>substance</u> is at least one of dimethoate, imidacloprid, fenpropidine, acephate and acetamiprid [is incorporated into the matrix].
- 15. (amended) Process according to [one or more of Claims] <u>Claim 1</u> [to 14] <u>or 2</u>, [<u>characterized</u> in that] <u>wherein in step a</u>) the <u>at least one</u> active substance [medium] is applied [in regular or irregular areal distribution onto] <u>to</u> at least one of the [matrix] layers [(1, 2), including its application, for example, in the form of patterns or stripes] <u>as a pattern</u>.

JONS Rec'd POTETTO 0 4 SEP 2001

Process for producing a sheetlike formulation comprising at least one single-layer active substance matrix

The invention relates to a process for producing a sheetlike formulation comprising at least one single-layer active substance matrix for the controlled release of active substance to the vicinity of the application site, the active substances being selected from crop protection agents, biocides, fertilizers, plant strengtheners, cosmetic active principles and fragrances.

Formulations, whose preparation is described below, are known from many areas of life for which the time- and quantity-controlled release of one or more active substances is required. Depending on the area of use, these formulations are applied in the form of sheetlike structures such as labels, strips, pouches, plasters or plaques to certain substrates or objects from which they develop their action. Typical fields of use for such products that may be mentioned here include, for example, pest control, for example, in vineyards or in forestry, or for crop protection, perfumery and cosmetology.

For reasons of cost, such formulations are wherever possible formulated so that the active substance is contained in a flat, single-layer polymer film from which in the course of the use the active substance is released to the site or the vicinity of application. In general, these formulations have a layered structure, comprising at least one matrix layer containing active substance and at least one support layer in contact with the matrix layer. These layers can be present in a "sandwich-like" form or are joined to one another at the sides and formed in the manner of a pouch. Depending on the field of use, they are usually also in fixed or detachable

combination with different functional layers such as, for example, control membranes or protective layers.

Processes for producing formulations of the type specified are known. A common feature of such preparation processes known from practice is that the formulation is first of all produced in the form of a strip-shaped web embracing the active substance matrix, then combined, if desired, with other layers, and separated into sections of a desired size by punching or cutting.

Because of the diversity of active substances applicable by means of these devices, and because of the different physicochemical properties of these active substances, the preparation of the single-layer active substance matrix represents the central step in such processes. Among the prior art processes for the preparation of such formulations, the dissolving or dispersing of the active substances in matrix material, usually a polymer solution or polymer melt, with subsequent drying, is probably the process of choice for the person skilled in the art. However, not all active substances can be processed in this way. The processing of volatile active substances proves to be particularly problematic, since the evaporation of the active substance during the preparation is almost impossible to control. And temperature-sensitive, so-called thermally labile active substances can be used only with restrictions, if at all, for systems requiring heat treatment during the manufacturing process.

For this reason, a variety of solutions have been worked out over the course of time to allow the preparation of the releasing matrices, especially for volatile and/or thermally labile active substances. For example, the principle of a depot is used, in which problematic active substances are introduced into a multi-part product in the

form of a discrete reservoir of active substance without thermal stress - for example, in a separate process step during preparation. Processes of this kind in which the preparation of matrix layers free from active substance and the introduction of the active substance into the product take place as temporally and spatially separate operating steps are known from the manufacture of numerous products but are very labour-intensive.

In accordance with the prior art, active substance can be incorporated into a matrix using a variety of techniques. For instance, the introduction of a flowable active substance medium under pressure, in addition to the pressurized insertion of a solid active substance body, and injection, belongs to known processes of incorporating active substance into solid matrices.

The processes mentioned have the common feature that active substance is applied initially to a support on which it remains only temporarily, since it migrates into the matrix layers that are in contact with it.

Constituents which prevent unwanted flow or escape and whose function is to halt the active substance temporarily are referred to in the technical literature using terms such as, for example, adsorption layer, active substance dispenser, active substance support, fixing aid, support material, and interlayer. Liquid-absorbing substrates suitable for this purpose include nonwoven, foam, paper or woven textile material.

In practice, however, known production processes are frequently associated with disadvantages, a selection of which is specified below, and, as a result, prove to be correspondingly complex:

The absorption capacity for liquid media is generally limited to a certain level, which is frequently a

limiting factor in loading the matrix with active substance.

- In order to obtain controlled, continuous release of active substance over prolonged periods of application and to rule out the possibility of unwanted side-effects caused by excessive release rates, an additional control layer is often necessary.
- The use of additional materials prolongs production and complicates it technically, since the individual layers and elements must first be manufactured separately from one another and then combined with one another in one or more subsequent steps.
- The use of additional elements may also impair the performance qualities of the product, especially the shear stability, since the incorporation of an active substance fixing means reduces the contact area between matrix layers which are to be united. In order to ensure, despite this, the desired structural integrity of the formulation, there is often a need for additional connecting areas, in the form, for example, of a peripheral adhesive edge, which result in an unwanted increase in the dimensions.

The embedding of additional support materials may, furthermore, adversely affect the flexibility and functionality of the product. This is particularly so if thicker layers are necessary as a result of a low absorptive capacity, e.g. the absorbency of the material. These disadvantages are extremely undesirable especially in the case of active substance patches applied to surfaces of plants.

The morphology of the higher plants, especially the low radius of curvature and the lack of evenness and cleanliness of surfaces of plants, indeed, imposes particularly stringent requirements on the flexibility and small dimensioning of such active substance formulations.

Added to this are the severe conditions borne by the surfaces of plants as a result of natural weather conditions at the site of application, which increases their susceptibility to a possible loss of cohesion.

Because of the disadvantages described above, multilayer or multipart formulations cannot be used for a number of applications on account, firstly, of their functionality. Secondly, the production costs resulting from the relatively high level of expenditure of material and manufacture limit the marketability and acceptance of such products. For price-sensitive markets and those where competition is intense, in particular, it would be desirable to be able to offer inexpensive products of comparatively simple construction for temperature-sensitive and volatile active substances as well.

The object of the invention is therefore to provide a process for producing sheetlike active substance formulations comprising a single-layer matrix, containing active substance, for the controllable release of active substances, including volatile and temperature-sensitive substances, which avoids the disadvantages of the processes known from the prior art.

This object is achieved in accordance with the invention by means of a production process in accordance with the process steps set out in the characterizing clause of Claim 1.

The invention is described in detail below.

A process is proposed in which the single-layer active substance matrix of the device is produced using at least two layers, identical in composition, of a base material in the following component steps which are separated from one another temporally and spatially (reference numerals in accordance with Fig. 1):

- a) provision of two prefabricable layers (1, 2), identical in composition, of a base material,
- application of active substance to at least one of the two layers (1, 2),
- c) placement of the two layers (1, 2) atop one another so as to enclose the active substance applied, and irreversible joining under pressure to form a laminate,
- d) storage of the laminate for predeterminable duration under defined conditions, with migration of the active substance into the base layers (1, 2), and connection at their interfaces to form a homogenous matrix featuring substantially uniform dispensation of active substance.

Active substances incorporated by this process into the matrix of the formulation include, for example, crop protection agents, biocides, fertilizers, plant strengtheners, cosmetic active principles and fragrances. The matrix layers can comprise one or more active substances.

"Biocides", according to the EC Biocides Directive, which has been in force since 14 May 1998, are substances or formulations which as intended possess the capacity to kill living organisms or at least to restrict them in their vital function. They are used, inter alia, as wood preservatives, disinfectants, process preservatives, insecticides and rodenticides.

The term "crop protection agents", which in the present specification is used synonymously with "pesticides", and also the term "plant strengtheners", are defined in § 2 of the Crop Protection Act applying in Germany.

The term "volatile substances" refers to substances having an effective vapour pressure even at room temperature. Examples that may be mentioned include insecticidal compounds such as dimethoate and acephate or insect pheromones such as Z,E-9,12-tetradecadienol and Z,E-9,12-tetradecadienol-1-yl acetate.

For the purposes of the present invention, the term "temperature-sensitive" or "thermally labile" substances refers to substances which decompose, or whose biological activity is impaired, at a temperature ≥ 50°C.

In the process of the invention, the single-layer active substance matrix comes about through direct introduction of active substance between identical matrix layers, which are subsequently united by pressure and stored for the purpose of "maturation". The principal advantage of this process lies in its avoidance of the use of additional active substance fixing aids, thereby considerably reducing the expenditure on material and manufacture and at the same time significantly improving the functional quality of the products produced in this way. As a manufacturing process which proceeds at room temperature, this process is of particular value for the production of devices comprising volatile and thermally labile substances.

Advantageous embodiments of the process according to the main claim can be inferred by the person skilled in the art from the features of the subclaims.

They relate, for example, to volumetric metering of the active substance, the operating pressure required when active substance is admitted into the matrix layer, further process parameters associated with the "maturation" of the matrix, the way in which the active substance is introduced and applied, the material basis of the matrix base

material, particular properties of the active substance, and its designations.

The principle of the invention is elucidated further in Fig. 1 on the basis of a process flowchart, while Fig. 2 shows the active substance matrix in section before and after storage.

In Fig. 1, (1) and (2) denote identical layers of the matrix base material, which are present as strip-shaped webs in the form of laminates on stock rolls (1a; 2a). Judiciously, both layers (1, 2) are provided on both sides with protective layers (3, 4), of which at least one (3) is detachable. The protective layers (3, 4) can consist of various materials such as paper, plastic and textiles, but must - by means, for example, of treatment with silicone - be rendered detachable. This is particularly important if the matrix layers (1, 2) are self-adhesive.

Prior to the application of the active substance, both prefabricated base material layers (1, 2) are exposed by the removable protective layers (3, 4) being peeled off and taken up by corresponding winders (5, 6).

Another configuration possibility for the process of the invention consists, for example, in the base material layers (1, 2) being provided only with one web, which is treated so as to repel the matrix on both sides, and being therefore present in the form of laminate rolls "wound in on themselves".

In the process of the invention, active substance is metered in the form of a flowable medium. The active substance medium is in a stock container (7) which is connected via a hose (8) to a metering station (10). The active substance medium is therefore located within a

closed system, which is of decisive importance in the case of volatile substances in particular.

Conveyance of the active substance medium into the metering station (10) is the function of a feed unit (9), which in the case of the process of the invention can be a hose pump, for example. However, it is also possible to use other kinds of pump suitable for metering, such as gear, screw, centrifugal or reciprocating pumps. It is essential for the purposes of the invention that the feed unit ensures pulseless conveying with a meterable conveyed quantity and constant pressure. A uniform conveyed flow is vital for uniform supplying of the metering station (10) with the active substance medium.

In the process of the invention, the metering station (10) consists, for example, of an active substance dispenser (11) and one or more applicator nozzles (12). The active substance dispenser (11) used in the process of the invention can comprise, for example, so-called NEMO Robo dispensers operating in accordance with the principle of rotating positive-displacement pumps. This functional principle of the dispenser is advantageous for the production process in that the conveyed amount of active substance medium is directly dependent on the rotor speed; it can be altered steplessly and adapted without problems to the requirements of the process. Different-sized constructions can be assigned different "metered-amount ranges"; as a result of the rotary mode of operation, the pressure exerted on the active substance medium remains constant.

Observing the uniform pressure, which is \leq 12 bar, is of essential importance to the invention in terms of metering accuracy.

A further advantage of this dispenser (11) lies in the possibility of reversing the conveying direction, which results in a short break in the thread. This prevents the accumulation of medium at the endpoints of the application and ensures uniform distribution of the active substance medium on the weblike matrix (14).

This is of particular importance for formulations in accordance with the process of the invention. Nonuniform distribution of the active substance between the base material layers (1, 2) would result in a nonhomogenous distribution of active substance in the matrix of the finished product and would, ultimately, have an adverse effect on the desired active substance release profile.

In this process, uniform distribution of the active substance formulation is the function of the applicator nozzles (12). Their number and arrangement are chosen so that the available matrix area is provided evenly with active substance medium. Judiciously, the application can be arranged in patterns such as stripes, dots, circles or other geometric shapes.

These patterns can be produced in either intermittent or rotary operating mode, preference being given to the latter mode since it generally permits higher production rates.

In the production process of the invention, active substances are metered in the form of a flowable medium whose viscosity can vary within wide limits but is at least 1000 mPa.s. Indeed, in the case of liquids of low viscosity, there can be unwanted flow of the medium on the matrix. This adverse effect would be intensified further in the course of the subsequent lamination under pressure. For establishing the desired minimum viscosity, viscosity-increasing additives such as, for example, AEROSIL ® or polymers are useful, which can be either of natural origin,

such as gelatine or derivatives of starch, for example, or of synthetic origin, such as polyacrylic acid, for example.

In order to establish the desired viscosity range it is judicious to thermally condition the application equipment.

In accordance with the invention, the laminating operation, which takes place directly after the metering of the active substance, can be performed at a pressure of between 2 and 10 bar. A particularly advantageous pressure range is that from 3 to 5 bar. In lamination, both base material layers (1, 2) are joined and irreversibly bonded under the effect of pressure. It is necessary to choose the laminating pressure such that the active substance medium does not emerge at the edges of the weblike matrix (14) and such that the interfaces of the base material layers (1, 2) are bonded inseparably.

A particularly advantageous possibility for configuring the production process of the invention consists in that base material layers (1, 2) and the matrix (14) formed from them have been made self-adhesive. This significantly facilitates the formation of the single-layer matrix (14) and additionally increases the shear stability of the finished product.

When the production process is carried out, the active substance medium can also have adhesive properties. A person skilled in the art can readily achieve this by means of tackifying additives, such as resins, for example.

The matrix-forming layers (1, 2) which are used in the process can consist of different materials. It is essential to the invention, however, that they are identical in terms of their composition and include a polymer or polymer mixture.

Suitable polymers in principle are all those which are able to take up and release active substances and which can be processed to films. The following groups may be mentioned as particularly suitable matrix polymers: ethylene-vinyl acetate copolymers, block copolymers, e.g. styrene/butadiene/styrene or styrene/isoprene/styrene, polyisobutylene, polyacrylates, polymethacrylates, polyvinyl esters, polyamide, polyesters, cellulose derivatives and silicones.

The selection of the polymer for base material layers (1, 2) is guided by the chemical and physical properties of the active substance.

Depending on the use of the formulation according to the invention, active substances from different groups can be used, such as crop protection agents, biocides, fertilizers, plant strengtheners, cosmetic active principles and fragrances, for example. The active substances, which following metering and lamination are firmly enclosed in the middle of the matrix (14), diffuse, in accordance with Figure 2 and in agreement with Fick's laws of diffusion, until they have attained a substantially uniform distribution within the matrix (14). This process, which is referred to as "maturation", is specific to the active substance and dependent on both time and temperature, and can be determined by the person skilled in the art. Advantageous embodiments comprise one or more of the active substances dimethoate, imidacloprid, fenpropidine, acephate and acetamiprid.

The duration of this phase of the production process of the invention can be shortened to a desired time by way of the nature of the operating parameters during storage of the laminate. It is important, however, that the storage temperature does not exceed the critical range for the active substance in question. The range 15-30°C and, in

particular, 20-24°C may be mentioned as a favourable storage temperature.

In a modification of the process of the invention, the maturation storage of the active substance matrix (14) can be followed by cooling at a temperature between 3 and 10°C. This has a positive effect on the cohesion and thus on the shear stability of the matrix (14) and is favourable to its mechanical strength in the case of punching or cutting.

In further process steps, the single-layer matrix produced in accordance with the invention, as shown in Figure 2, is combined with other functional layers and then processed to give individual plaster-like formulations.

The invention is described below with reference to an example, which represents a preferred embodiment of the invention.

Example

In Fig. 1, the web (1a) is a laminate whose width, for example, is 54 mm and which, as viewed from bottom to top, consists of a 36 μm thick PET support film, a 125 g/m² base material layer made of polyacrylate, and a siliconized protective layer (95 g/m² paper). The second web of laminate (2a) is also 54 mm wide and comprises, as viewed from bottom to top, 95 g/m² siliconized paper, a 125 g/m² base material layer, identical with that of the laminate (1a), and an 80 μm thick siliconized PE protective film. Before metering, the protective layers (3, 4) of the two laminates (1a, 2a) are peeled off and taken up by the winders (5) and (6). The remaining laminates are taken into the laminating unit (13) such that the weblike base material layer of the laminate (1a) is congruent with the web lying on the counter-roller.

The active substance formulation, which has a viscosity of 1100 mPa.s and comprises 52.46% by weight dimethoate, 34.76% by weight N-methylpyrrolidone and 12.78% by weight colloidal silica, is metered continuously onto the centre of the base material strip (polyacrylate film) of the laminate (1a) by means of three applicator nozzles (12), metering taking place at a pump speed of 850 rpm and a machine running speed of 20 m/min. The amounts of active substance applied are 0.386 g per 0.64 m of the laminate (1a). Directly after the active substance medium has been metered, it is covered with the second base material layer (2a). This operation, referred to as laminating, is carried out under a pressure of 3 bar.

The resultant matrix laminate (14) containing dimethoate is stored at 20°C for 14 days for the purpose of maturation and subsequently at 6°C for 24 hours.

The finishing of the formulation, by uniting the matrix (14) with a final cover, and the subsequent processing, take place in a punching and processing unit.

CLAIMS

- 1. Process for producing a sheetlike formulation comprising at least one single-layer active substance matrix for the controlled release of active substance to the vicinity of the application site, the active substances being selected from crop protection agents, biocides, fertilizers, plant strengtheners, cosmetic active principles and fragrances, characterized in that the matrix is produced using at least two layers (1, 2), identical in composition, of a base material in the following temporally and spatially separate steps:
 - a) provision of two prefabricable layers (1,2), identical in composition, of a base material,
 - b) application of active substance to at least one of the two layers (1,2),
 - c) placement of the two layers (1,2) atop one another so as to enclose the active substance applied, and irreversible joining under pressure to form a laminate,
 - d) storage of the laminate for predeterminable duration under defined conditions, with migration of the active substance into the base layers (1,2), and connection at their interfaces to form a homogenous matrix featuring substantially uniform dispensation of active substance.
- 2. Process according to Claim 1, <u>characterized</u> in that volumetric metering of the active substance medium is performed by admitting it at a pressure ≤ 12 bar.

- 3. Process according to Claim 1 or 2, characterized in that the joining of the base material layers (1, 2) is performed under a laminating pressure of between 2 and 10 bar, preferably between 3 and 5 bar.
- 4. Process according to one or more of Claims 1 to 3,

 characterized in that the storage of the active
 substance matrix (14) is performed at a temperature of
 between 15 and 30°C, preferably between 20-24°C, with
 a duration of at least 48 hours.
- 5. Process according to one or more of Claims 1 to 4,

 <u>characterized</u> in that active substance is applied in
 the form of a flowable medium having a viscosity of at
 least 1000 mPa.s.
- 6. Process according to Claim 5, <u>characterized</u> in that active substance medium is applied with a fraction of auxiliaries.
- 7. Process according to one or more of Claims 1 to 6,

 <u>characterized</u> in that the application of active
 substance is performed continuously or intermittently.
- 8. Process according to one or more of Claims 1 to 7,

 <u>characterized</u> in that the active substance matrix (14)
 is made self-adhesive.
- 9. Process according to one or more of Claims 1 to 8,

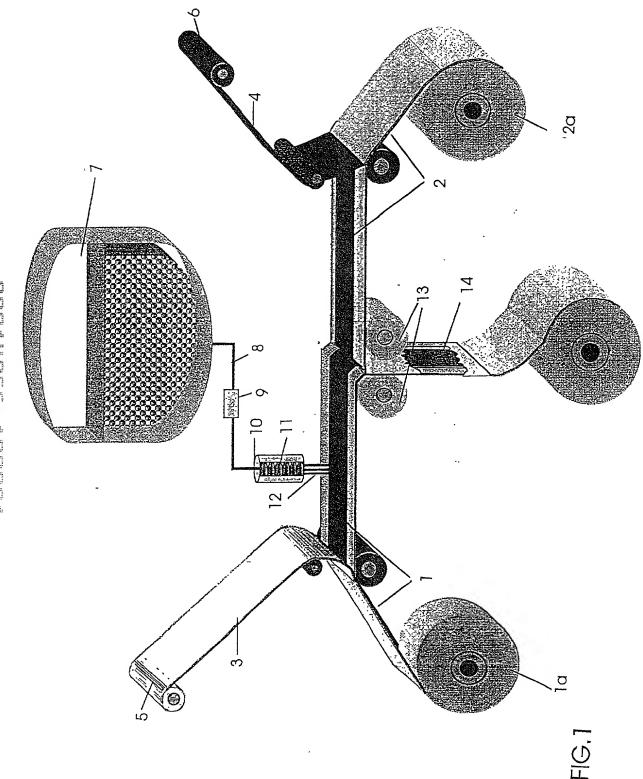
 <u>characterized</u> in that volatile or thermally labile
 active substances are incorporated between the base
 material layers (1, 2) of the matrix.
- 10. Process according to one or more of Claims 1 to 9, characterized in that at least one polymer from one of

the following groups is selected as the matrix base material: ethylene-vinyl acetate copolymer, block copolymer, e.g. styrene/butadiene/styrene or styrene/isoprene/styrene, polyisobutylene, polyacrylates, polymethacrylates, polyvinyl esters, polyamide, polyesters, cellulose derivatives and silicones.

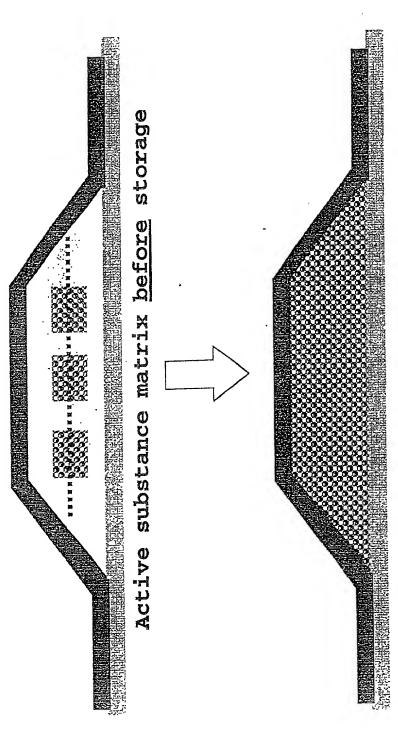
- 11. Process according to one or more of Claims 1 to 10,

 <u>characterized</u> in that an active substance medium is used which comprises tackifying substances.
- 12. Process according to one or more of Claims 1 to 11, characterized in that an active substance is used which is a mixture of Z,E-9,12-tetradecadienol and Z,E-9,12-tetradecadien-1-yl acetate.
- 13. Process according to one or more of Claims 1 to 12,

 characterized in that the matrix layers comprise one
 or more active substances.
- 14. Process according to one or more of Claims 1 to 13, characterized in that at least one of the active substances dimethoate, imidacloprid, fenpropidine, acephate and acetamiprid is incorporated into the matrix.
- 15. Process according to one or more of Claims 1 to 14, characterized in that the active substance medium is applied in regular or irregular areal distribution onto at least one of the matrix layers (1, 2), including its application, for example, in the form of patterns or stripes.



The state of the s



Active substance matrix after storage

FIG.

Logical Control

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY

(Includes Reference to PCT International Applications)

Attorney's Docket Number

F-7104

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

METI	<u>IOD FOR PRODUCING A FLAT PREPARATION WITH AT</u>	LEAST ONE
SINGI	E-LAYER INGREDIENT MATRIX	· .
the specifi	cation of which (check only one item below):	
[]	is attached hereto.	
_ []·	was filed as United States application	
	Serial No.	
il constant	on	
	and was amended	
iur 1 ¹⁰ g	on	(if applicable).
	was filed as PCT international application	
1 1	Number <u>PCT/EP00/01438</u>	
	on February 22, 2000	
	and was amended under PCT Article 19	
	on	(if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed:

Country (if PCT indicate "PCT")	Application Number	Date of Filing (day, month, year)	Priority Claimed Under 35 USC 119
Jermany	199 09 493.4	4, March 1999	[X]Yes []No
			[]Yes []No
			[]Yes []No
			[]Yes []No
		·	[]Yes []No

The Hand And Build Boy of

COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY (Continued)

(Includes Reference to PCT International Applications)

Attorney's Docket Number

F-7104

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

RIOR U.S. APPLICATIONS OF 5 U.S.C. 120.	r pct international ap	PLICATIONS DESIGNATING	THE U.S. FOR BEN	EFIT UNDER	
5 0.0.0.	U.S. APPLICATIONS		s	TATUS (Check O	nc)
U.S. Application Number	U. S.)	Filing Date	Patented	Pending	Abandone
			_		
PCT API	PLICATIONS DESIGNATING	THE U.S.			
PCT Application No.	PCT Filing Date	U.S. Serial Numbers Assigned (if any)			
J.					
	Reg. No. 20,456	nd Trademark Office com Herbert F. Ruschma Marvin Turken Alfred D'Andrea	inn Reg. N	o. <u>35,34</u> 1 o. 18,330	
Send Correspondence To	o: <u>Jordan and Har</u> 122 Bast 42nd New York, New	Street	Direct	Telephone Ca C. Bruce Ha (212) 986-	mburg
nd belief are believed to be	e true; and further that the nishable by fine or impris	my own knowledge are tress statements were made sonment, or both, under sector the validity of the appl	with the knowled ction 1001 of Tit	dge that willfu le 18 of the Un	l false state: tited States
nd belief are believed to be nd the like so made are put nd that such willful false Full Name of Sole or First Investor	e true; and further that the nishable by fine or impris	ese statements were made sonment, or both, under se	with the knowled ction 1001 of Tit	dge that willfulle 18 of the Unatent issuing to	l false stater tited States (hereon.
nd belief are believed to be	e true; and further that the nishable by fine or impris	se statements were made sonment, or both, under set ze the validity of the appl	with the knowled ction 1001 of Tit	dge that willfulle 18 of the Unatent issuing to	l false stater tited States (
nd belief are believed to be nd the like so made are put nd that such willful false Full Name of Sole or First Inventor Michael ROREGER Residence Neuwied, Germany	e true; and further that the nishable by fine or impris	se statements were made sonment, or both, under set ze the validity of the appl	with the knowled ction 1001 of Tit	dge that willfulle 18 of the Unatent issuing to	l false stater tited States (hereon.
nd belief are believed to be nd the like so made are pur nd that such willful false Full Name of Sole or First Inventor Michael ROREGER Residence	e true; and further that the nishable by fine or imprisoratements may jeopard	ese statements were made sonment, or both, under seize the validity of the appl	with the knowled ction 1001 of Tit	dge that willfulle 18 of the Unatent issuing to	l false stater tited States (hereon.
nd belief are believed to be nd the like so made are pure nd that such willful false Full Name of Sole or First Inventor Michael ROREGER Residence Neuwied, Germany Fract Office Address	e true; and further that the nishable by fine or imprisoratements may jeopard	ese statements were made sonment, or both, under seize the validity of the appl	with the knowled ction 1001 of Tit	dge that willfulle 18 of the Unatent issuing to	l false states uited States hereon.
nd belief are believed to be nd the like so made are pur nd that such willful false Full Name of Sole or First Inventor Michael ROREGER Residence Neuwied, Germany Fest Office Address Pestalozzi-Strasse 38, D	e true; and further that the nishable by fine or imprisstatements may jeopard	ese statements were made sonment, or both, under seize the validity of the appl	with the knowlection 1001 of Tit lication or any p	dge that willfulle 18 of the Unatent issuing to	l false states (hereon.
nd belief are believed to be nd the like so made are pure nd that such willful false Full Name of Sole or First Inventor Michael ROREGER Residence Neuwied, Germany Fest Office Address Pestalozzi-Strasse 38, D	e true; and further that the nishable by fine or imprisstatements may jeopard	ese statements were made sonment, or both, under seize the validity of the appl	with the knowlection 1001 of Tit lication or any p	pare 22.08 Citizenship Germany	l false states (hereon.